# Status Update on the High Precision Isotopic Measurements on High Burnup LWR Fuel in 2020

**Spent Fuel and Waste Disposition** 

Prepared for US Department of Energy Spent Fuel and Waste Science and Technology

Oak Ridge National Laboratory Bruce Bevard, Joseph Giaquinto, Germina Ilas, Ian Gauld

> September 30, 2020 M4SF-20OR010308091 ORNL/SPR-2020/1762

#### DISCLAIMER

This information was prepared as an account of work sponsored by an agency of the U.S. Government. Neither the U.S. Government nor any agency thereof, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness, of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof.

This is a technical paper that does not take into account contractual limitations or obligations under the Standard Contract for Disposal of Spent Nuclear Fuel and/or High-Level Radioactive Waste (Standard Contract) (10 CFR Part 961). For example, under the provisions of the Standard Contract, spent nuclear fuel in multi-assembly canisters is not an acceptable waste form, absent a mutually agreed to contract amendment.

To the extent discussions or recommendations in this paper conflict with the provisions of the Standard Contract, the Standard Contract governs the obligations of the parties, and this paper in no manner supersedes, overrides, or amends the Standard Contract.

This paper reflects technical work which could support future decision making by DOE. No inferences should be drawn from this paper regarding future actions by DOE, which are limited both by the terms of the Standard Contract and a lack of Congressional appropriations for the Department to fulfill its obligations under the Nuclear Waste Policy Act including licensing and construction of a spent nuclear fuel repository.

Reactor & Nuclear Systems Division

## STATUS UPDATE ON THE HIGH PRECISION ISOTOPIC MEASUREMENTS ON HIGH BURNUP LWR FUEL IN 2020

Bruce B. Bevard Joseph Giaquinto Germina Ilas Ian Gauld

September 2020

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831-6283
managed by
UT-BATTELLE, LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

## **CONTENTS**

ACI	RONYMS	iv
ABS	STRACT	
	INTRODUCTION	
	DATA GAPS FOR PWR AND BWR SNF	
3.	ORNL'S HIGH PRECISION RADIOCHEMICAL ANALYSIS PROGRAM	
	3.1 PWR DATA COLLECTED TO DATE	6
	3.2 SIMULATED DATA FOR PWR SAMPLES	
4.	BWR SAMPLE INFORMATION	
5.	SUMMARY	<u>(</u>
6.	REFERENCES	10
APF	PENDIX A. PWR SAMPLE INFORMATION	A-1
APF	PENDIX B. BWR SAMPLE INFORMATION	B-1
	PENDIX C. ISOTOPES TO BE ANALYZED	

#### **ACRONYMS**

ASTM American Society for Testing and Materials

BWR boiling water reactor
C/E calculated-to-experimental
CRM certified reference materials
DOE US Department of Energy

DPC dual-purpose (storage and transportation) canisters

FY fiscal year

GUM Guide to the Expression of Uncertainty in Measurement

GWd/MTU gigawatt-days per metric ton of uranium

HBU High burnup

HP-RCA high-precision radiochemical analysis IAEA International Atomic Energy Agency

ICPMS inductively coupled plasma mass spectrometry

ID isotope dilution

ISO/IEC International Organization for Standardization/International Electrotechnical Commission

LCS laboratory control sample

MC-ICPMS multi-collector inductively coupled plasma mass spectrometry MC-TIMS multi-collector thermal ionization mass spectrometry

MWd/tU megawatt-day per metric ton uranium

NBL New Brunswick Laboratory
NDE nondestructive examination
NE Office of Nuclear Energy
NEA Nuclear Energy Agency

NIST National Institute of Standards and Technology

ORNL Oak Ridge National Laboratory
PWR pressurized water reactor
RCA radiochemical assay
R&D research and development

SFWD Spent Fuel and Waste Disposition

SFWST Spent Fuel and Waste Science and Technology

SNF spent nuclear fuel

TIMS thermal ionization mass spectrometry

WRM working reference material

Wt weight percent

#### **ABSTRACT**

The US Department of Energy (DOE) Office of Nuclear Energy (NE) is currently investigating the feasibility of directly disposing dual-purpose (storage and transportation) canisters (DPCs) in a spent nuclear fuel (SNF) repository. Criticality during the repository performance period (10,000 years or more) is one of the major concerns related to direct disposal of DPCs, specifically as the system undergoes degradation in the repository environment and timeframe. Oak Ridge National Laboratory (ORNL) is developing an as-loaded criticality analysis methodology using full (actinides + fission products) burnup credit that exploits the inherent criticality margin associated with actual canister-specific loading configuration. Burnup credit criticality analysis requires validation of the depletion/decay codes used to generate the burned isotopic inventory of an assembly by comparing the code-predicted inventory with experimentally determined isotopic data. Currently, isotopic measurement data for boiling water reactors (BWR) SNF are limited, and additional measurements will be highly beneficial for BWR burnup credit analysis, which is essential to demonstrate disposability of BWR DPCs. Moreover, new pressurized water reactor (PWR) samples of isotopic measurements will expand the PWR sample population and consequently will greatly improve sample statistics. In turn, this will reduce uncertainty in the computational determination of the isotopic composition of commercial SNF and eliminate additional penalties currently used for lack of data.

As part of the isotopic analysis task, eight diverse, high burnup (HBU) samples from several PWR rods are being dissolved and isotopically analyzed to provide high-quality measurement data for PWR SNF to reducing the uncertainties associated with PWR isotopic depletion validation. The isotopics of interest focus on nuclides important to burnup credit, shielding, and decay heat in PWR SNF. Additionally, eight high burnup BWR fuel samples from the Limerick nuclear power plant have been identified for measurement to support reducing uncertainties associated with BWR isotopic depletion validation.

#### 1. INTRODUCTION

This report documents work performed under the Spent Fuel and Waste Disposition, Spent Fuel and Waste Science and Technology program for the US Department of Energy (DOE) Office of Nuclear Energy (NE). This work was performed to fulfill Level 4 Milestone M4SF-20OR010308091, "Status Update on the High Precision Isotopic Measurements on High Burnup LWR Fuel in 2020," within work package SF-20OR01030809.

The US Department of Energy (DOE) Office of Nuclear Energy (NE) is currently investigating the feasibility of directly disposing dual-purpose (storage and transportation) canisters (DPCs) in a spent nuclear fuel (SNF) repository. Criticality during the repository performance period (10,000 years or more) is one of the major concerns related to direct disposal of DPCs, specifically as the system undergoes degradation in the repository environment and timeframe. Oak Ridge National Laboratory (ORNL) is developing an as-loaded criticality analysis methodology using full (actinides + fission products) burnup credit that exploits the inherent criticality margin associated with actual canister specific loading configuration. Burnup credit criticality analysis requires validation of the depletion/decay codes used to generate the burned isotopic inventory of an assembly by comparing the code-predicted inventory with experimentally determined isotopic data. Currently, isotopic measurement data for boiling water reactor (BWR) SNF are limited, and additional measurements will be highly beneficial for BWR burnup credit analysis, which is essential to show disposability of BWR DPCs. Moreover, new pressurized water reactor (PWR) samples of isotopic measurements will expand the PWR sample population and consequently will greatly improve sample statistics. In turn, this will reduce uncertainty in the

computational determination of the isotopic composition of commercial SNF and eliminate additional penalties currently used for lack of data.

High-quality radiochemical assay (RCA) data are important for evaluating uncertainties in SNF safety analyses, including burnup credit, decay heat, neutron and gamma sources, or waste management applications. In particular, they provide one means for determining uncertainties in integral quantities important to safety, such as decay heat or SNF reactivity. Direct measurements of such integral quantities can be expensive or impractical for covering the multitude of existing fuel designs, operating conditions, and specific application purposes. However, as these integral quantities are mainly driven by the nuclide composition in SNF following irradiation and the decay time after discharge, measured nuclide compositions can serve as an indirect way to determine uncertainties associated with code predictions of these quantities. The minimum uncertainties associated with these types of measurements are 2-3% at a 95% confidence level. Isotope ratio measurements using multi-collector thermal ionization mass spectrometry (MC-TIMS) and multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS) are among the most accurate mass spectrometry techniques; they can reach accuracies of 0.1– 0.4% for major isotopes when used in tandem with isotope dilution (ID) as a calibration technique using high precision certified reference materials. It is important to note that for existing data, the reported measurement uncertainties, including their value and significance, differ greatly among measurement laboratories. There is a general lack of consistency in reporting uncertainties—overall uncertainty vs. spectrometry-only uncertainty across laboratories and different experimental programs.

DOE-NE, in partnership with the Electric Power Research Institute, developed the High Burnup (HBU) Spent Fuel Data Project to perform a large-scale demonstration and laboratory-scale testing of HBU PWR fuels (exceeding 45 gigawatt-days per metric ton of uranium [GWd/MTU]). Under this project, 25 sister rods—which are rods of the same design, similar power histories, and other similar characteristics—were removed from assemblies at the North Anna Nuclear Power Station and sent to ORNL in January 2016. ORNL performed detailed nondestructive examination (NDE) on all 25 rods. The NDE consisted of visual examinations, gamma scanning, profilometry and rod length measurements, and eddy current examinations. As part of the isotopic analysis task, eight diverse, HBU samples from several of the sister rods are being dissolved and isotopically analyzed to provide high-quality measurement data for PWR spent fuel to reduce uncertainties associated with PWR isotopic depletion validation. The isotopics of interest focus on nuclides important to burnup credit, shielding, and decay heat in PWR SNF (Appendix C).

BWR fuel comprises approximately 56% [1] of the total fuel assemblies currently in storage at nuclear power plants in the United States. BWR nuclear fuel and cladding designs and manufacturing are significantly different from PWRs. Differences include the following items, and they affect the isotopic concentrations of discharged BWR SNF:

- BWR fuel pellets are larger than PWR pellets.
- Variations of Zircaloy-2 (including liners) are used instead of the Zircaloy-4 cladding materials used in PWRs.
- Clad manufacturing and stress-relief processes are different between PWRs and BWRs.
- The fuel rod dimensions are different because larger rod diameters and thicker cladding are used in BWRs.
- BWR fuel typically has lower internal rod pressures and sees vastly different operating conditions than PWR fuel (i.e., two-phase flow).

- BWR assemblies are "canned," meaning each assembly is surrounded by a metal fuel channel.
- BWR cladding is often composed of an inner pure Zr liner that has widely different mechanical properties than the Zircaloy-2 alloy, and it exhibits a stronger affinity for hydrogen.
- The BWR SNF generally has more total hydrogen in the cladding/liner than typical PWR fuel.

To date, Spent Fuel and Waste Disposition (SFWD) program efforts have focused on the PWR fuel that is part of the sister rod test program. HBU SNF rod segments from the BWR Limerick nuclear power station have been identified in the ORNL hot cells. These fuel segments have been used in previous DOE programs to support cladding behavior and fuel rod performance studies. Fuel rod segments from rods J4 and J6 of the GE11 ( $9 \times 9$ ) assembly YJ1433 are available for destructive analysis measurements. The fuel segments provide the opportunity to obtain high-precision measurement data for sample burnups in the range 40—65 GWd/tU. This experimental program will obtain up to eight BWR rod samples, along with the associated operating data, and they will be isotopically analyzed at ORNL to support closing the BWR isotopic data gap.

#### 2. DATA GAPS FOR PWR AND BWR SNF

Quantifying and evaluating the bias and uncertainties in code predictions of SNF compositions is essential for validating the accuracy of the codes and nuclear data used for PWR and BWR safety and licensing calculations. Determination of the bias and uncertainties in code predictions of isotopic compositions is a continuous process. These values must be reassessed to keep pace with continuous changes in the characteristics of SNF currently being discharged or planned for discharge from commercial reactors in the future. The modern fuels are characterized by higher burnups, higher enrichments, complex and heterogeneous assembly designs, and improved reactor operation. To cover the broad fuel characteristics of relevance to SNF applications, a comprehensive experimental database is needed. For independent validation of depletion methods and data, calculated isotopic predictions have traditionally been compared to RCA data to determine biases and uncertainties for each isotope considered in the safety evaluation. The calculational bias is defined as the average measured-tocalculated ratio for a number of comparisons for a given isotope. The uncertainty in the bias is the product of the standard deviation of the bias and a tolerance factor corresponding to a desired confidence level. The uncertainty is typically accounted for at a 95% confidence level and reflects the variance of the predicted bias and the number of assay measurements available. For isotopes with relatively few measurements, the uncertainty can be large--decreasing the benefits of high-fidelity depletion analyses. Accurate predictions of the isotopic compositions are the foundational element for all applications that involve spent fuel (e.g., thermal analyses, radiation dose analyses, and criticality safety).

Nuclides of high importance to safety applications involving spent nuclear fuel have been previously identified and discussed [2–6]. Table 1 presents a list of the nuclides [2] important to burnup credit, decay heat, disposal, and other waste management applications.

Nuclide	Half life	Burnup credit	Decay heat	Waste management
<sup>79</sup> Se	$2.95 \times 10^5$ years			•
<sup>95</sup> Mo	Stable			
<sup>90</sup> Sr/ <sup>90</sup> Y	28.9 years			
<sup>99</sup> Tc	$2.11 \times 10^{5}$	•		•
<sup>101</sup> Ru	Stable			

Table 1. Nuclides important to SNF safety applications [2]

<sup>106</sup> Ru	371.6 days		
<sup>103</sup> Rh	Stable		
<sup>109</sup> Ag <sup>125</sup> Sb	Stable		
<sup>125</sup> Sb	2.76 years		
129I	$1.6 \times 10^7$ years		
<sup>133</sup> Cs	Stable		

Table 1. Nuclides important to SNF safety applications [2] (continued).

Nuclide	Half life	Burnup credit	Decat heat	Waste management
<sup>134</sup> Cs	2.06 years			
<sup>135</sup> Cs	$2.3 \times 10^6$ years			
<sup>137</sup> Cs/ <sup>137</sup> Ba	30.0 years			
$^{139}$ La $^a$	Stable			
<sup>143</sup> Nd	Stable	•		
<sup>145</sup> Nd	Stable	•		
$^{148}\mathrm{Nd}^a$	Stable			
<sup>144</sup> Ce/ <sup>144</sup> Pr <sup>a</sup>	284.9 days			
<sup>155</sup> Gd	Stable			
<sup>147</sup> Sm	1.06 × 10 <sup>11</sup> years			
<sup>149</sup> Sm	Stable			
<sup>150</sup> Sm	Stable			
<sup>151</sup> Sm	90 years			
<sup>152</sup> Sm	Stable			
<sup>151</sup> Eu	Stable			
<sup>153</sup> Eu	Stable	•		
<sup>154</sup> Eu	8.59 years		•	
<sup>155</sup> Eu <sup>b</sup>	4.75 years	•	•	
<sup>234</sup> U	2.45 × 10 <sup>5</sup> years			
235U	7.04 × 10 <sup>8</sup> years	•		•
236U	$2.34 \times 10^7$ years	•		•
<sup>238</sup> U	4.47 × 10 <sup>9</sup> years	•		
<sup>237</sup> Np	2.14 × 10 <sup>6</sup> years	•		
<sup>238</sup> Pu	87.71 years	•	•	
<sup>239</sup> Pu	$2.41 \times 10^{4} \text{ years}$	•	•	
<sup>240</sup> Pu	$6.56 \times 10^3$ years		•	
<sup>241</sup> Pu	14.29 years	•		
<sup>242</sup> Pu	$3.75 \times 10^5$ years	•		
<sup>241</sup> Am	433 years			
<sup>243</sup> Am	7,370 years	•		•
<sup>242</sup> Cm	162.8 days		•	
<sup>243</sup> Cm <sup>c</sup>	29.1 years	•		
<sup>244</sup> Cm	18.1 years		•	
<sup>245</sup> Cm	$8.5 \times 10^3$ years	•		•
<sup>246</sup> Cm <sup>c</sup>	18.1 years			`

The 28 nuclides (12 actinides and 16 fission products) with specific importance for burnup credit for storage and transportation [6,7] are listed in Table 2. Their relative importance to SNF reactivity varies with burnup, cooling time, enrichment, and assembly design. Six of 16 fission products (143Nd, 149Sm, 103Rh, 151Sm, 133Cs, and 155Gd) account for ~75% of the fission product reactivity's worth and ~20% of the total reactivity's worth in typical SNF [5]. High-quality measurement data for the nuclides listed in Table 1 above and Table 2 below are critical to validate the codes used for calculating the nuclide inventories in spent fuel, which is essential for spent fuel storage, transportation, and final disposal.

Table 2. Nuclides important to burnup credit for storage and transportation [6,7].

<sup>234</sup> U	<sup>235</sup> U	<sup>236</sup> U	<sup>238</sup> U	<sup>237</sup> Np	<sup>238</sup> Pu
<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>241</sup> Am	<sup>243</sup> Am
<sup>95</sup> Mo	<sup>99</sup> Tc	<sup>101</sup> Ru	<sup>103</sup> Rh	<sup>109</sup> Ag	<sup>133</sup> Cs
<sup>143</sup> Nd	<sup>145</sup> Nd	<sup>147</sup> Sm	<sup>149</sup> Sm	<sup>150</sup> Sm	<sup>151</sup> Sm
<sup>152</sup> Sm	<sup>151</sup> Eu	<sup>153</sup> Eu	<sup>155</sup> Gd	=	-

Previous studies [7] emphasized that high-quality RCA data for the two major actinides—<sup>235</sup>U and <sup>239</sup>Pu—are critical, as they drive the generation of higher actinides and fission products. Addition of new samples for HBU fuel will increase the number of samples in the burnup range of > 40 GWd/MTU, for which large uncertainties were noted in the major actinide <sup>239</sup>Pu, other Pu nuclides, and fission products [6,7]. The validation for burnup credit will also benefit the availability of high-quality data for the six important fission products (see Table 2), especially for <sup>155</sup>Gd, for which existing experimental data are scarce, with large measurement uncertainties [6,7].

The current publicly available BWR measurement data include 77 samples [6], with over 80% of these samples from Fukushima Daini BWRs operated in Japan. Over 40 of the 77 samples are for fuel selected from  $8 \times 8$  or  $6 \times 6$  older assembly design. Many of the samples do not include all the nuclides important for burnup credit (Table 2). There is a real benefit in adding new, high-quality BWR RCA data to existing data to impact the reduction of uncertainty in the calculated nuclide inventory.

#### 3. ORNL'S HIGH PRECISION RADIOCHEMICAL ANALYSIS PROGRAM

ORNL has developed and qualified a comprehensive analytical protocol for the high-precision characterization of key fission product and actinide isotopes in select specimens from the HBU fuels. Combining its decades of nuclear material characterization expertise performed for SNF, isotope production, nuclear forensics, and reference materials, ORNL Nuclear Analytical Chemistry has established expertise and capabilities for high-precision hot cell dissolutions, high-resolution chemical separations, and high-precision isotopic measurements that will offer results with unprecedented measurement precision and will greatly improve upon existing data sets.

<sup>&</sup>lt;sup>a</sup> Nuclides used as burnup indicators

<sup>&</sup>lt;sup>b</sup> Important not directly, but as parent nuclide of <sup>155</sup>Gd

<sup>&</sup>lt;sup>c</sup> Important for very high burnup.

A key to ORNL's capabilities in high-precision radiochemical analysis (HP-RCA) is the laboratory's state-of-the-art MC-ICPMS system for radioactive sample types. This instrument is capable of providing isotope ratio measurements with relative uncertainties on the order of 0.1%. This new, high-performing measurement instrument has been used for measurements on a safeguards project and will be used to perform the RCA isotope ratio measurements for these LWR samples.

#### 3.1 PWR DATA COLLECTED TO DATE

Eleven PWR samples have been selected from the sister rods: three for chemical determination of burnup, and eight for full-isotopic analysis (see Appendix A). Appendix C presents the isotopes included for the full-isotopic measurements. The three samples selected for burnup-only measurements have operator-estimated burnups of 42.5, 59.1, and 61.2 GWd/tU. The eight samples planned for full-isotopic analysis cover an HBU range of 53.3–63.5 GWd/tU.

Resolutions

High Resolution Chemical Separations

High Precision Measurement Methodologies

Figure 1. Flow diagram of ORNL HP-RCA protocols

To date, Nuclear Analytical Chemistry has completed the chemical determination of burnup on three fuel specimens according to ASTM E321, Standard Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method) [8]. A comprehensive sample analysis plan was developed for the HP-RCA measurements, along with a working reference material (WRM) using National Institute of Standards and Technology (NIST)- and New Brunswick Laboratory (NBL)-certified reference materials (CRM). All ID spikes were prepared to be used as calibrants, and the hot cell dissolutions on four fuel specimens for HP-RCA were completed.

The WRM will be processed in parallel with the fuels and used as a laboratory control sample (LCS) to document performance of the analytical protocol. The ID spikes were created using high-precision protocols and will allow for high-precision quantifications in the unknowns. It should be noted that for the majority of the analytes to be measured, there are no ID spikes available, so the spikes used for this project were created using enriched isotopes procured from DOE's Isotope Business Offices, with traceability established to NIST standards performed using ORNL high-precision measurement protocols.

Calculations with measurement protocols compliant with the Guide to the Expression of Uncertainty in Measurement (GUM, ISO/IEC GUIDE 98-3:2008) have been established for all nuclides listed in Appendix C to report total uncertainties for the results. A further study of the optimal separation technique is needed for <sup>79</sup>Se.

Additionally, as a proof of concept, the Nd fractions from the three burnup samples were analyzed using ID MC-ICPMS. Comparison between the two data sets showed an improvement in relative precision for elemental Nd from  $\pm$ 0.5 % to  $\pm$ 0.5 % and its isotopic abundances from  $\pm$ 0.08 % to  $\pm$ 0.005 %. These are 2-sigma total uncertainties.

#### 3.2 SIMULATED DATA FOR PWR SAMPLES

Preliminary depletion and decay simulations have been performed for the eight PWR samples selected for full isotopic analysis. The calculated nuclide inventories were needed as a basis to quantify the needed isotopic dilution spikes used as calibrants in the planned measurements. These simulations were performed using the Origami interface of the ORIGEN isotopic depletion and decay code in the ORNL SCALE 6.2.4 nuclear analysis code system [9]. Cross section libraries representative of a 17 × 17 PWR assembly design have been used with Origami. Selected nuclides calculated for the 63.5GWd/tU sample at a cooling time corresponding to August 2020 are listed as an example in Table 3.

Table 3. Isotopic composition for 63.5GWd/tU PWR sample calculated with Origami

Isotope	g/tU initial
u234	2.32E+02
u235	4.03E+03
u236	6.09E+03
u238	9.10E+05
np237	8.97E+02
pu238	5.21E+02
pu239	6.12E+03
pu240	3.43E+03
pu241	7.39E+02
pu242	1.38E+03
am241	1.28E+03
am242m	8.99E-01
am243	3.78E+02
cm242	2.34E-03
cm243	9.76E-01
cm244	1.17E+02
cm245	2.07E+01
cm246	4.48E+00
cm247	8.75E-02
se79	8.77E+00
sr90	5.57E+02
ru101	1.44E+03
ru106	2.68E-04
rh103	7.79E+02
sb125	9.61E-02

Isotope	g/tU initial
i129	3.06E+02
cs133	1.92E+03
cs134	3.50E-01
cs135	7.13E+02
cs137	1.43E+03
nd142	6.71E+01
nd143	1.17E+03
nd144	2.73E+03
nd145	1.14E+03
nd146	1.42E+03
nd148	7.05E+02
nd150	3.51E+02
ce144	5.74E-06
pm147	1.03E+00
sm147	3.46E+02
sm149	4.02E+00
sm150	5.57E+02
sm151	1.40E+01
sm152	1.56E+02
eu151	2.39E+00
eu152	3.30E-03
eu153	2.12E+02
eu154	1.06E+01
eu155	8.93E-01
gd155	1.67E+01

Higher fidelity depletion and decay simulations were performed for the three samples used for burnuponly measurements. These simulations were performed with the TRITON depletion capability in SCALE, which iteratively couples ORIGEN with a neutron transport solver. The simulations used 2D models of the fuel assembly, assembly-specific design data, and operating history data. The 2D assembly model used for depletion simulations is illustrated in Figure 2.

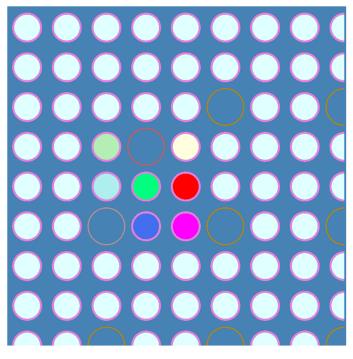


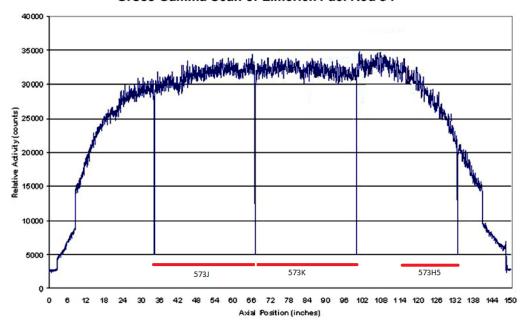
Figure 2. Illustration of depletion model (1/4 assembly).

#### 4. BWR SAMPLE INFORMATION

Eight samples from Limerick assembly YJ1433 rods J4 and J6 have been identified for measurement. The majority of fuel segments are from rod J4, and six samples are planned for measurement. Two additional samples from rod J6 (symmetric assembly location) are planned.

All fuel rod samples have an initial enrichment of 3.95 wt% <sup>235</sup>U. Sample locations are selected to provide the greatest range of burnup values within the same J4 fuel rod. In addition, sample locations are selected to provide a wide range of coolant void fraction values with a significant effect on the nuclide evolution in the fuel. The gross gamma scan of rod J4 is shown in Figure 3. The available rod segments are indicated. The maximum rod burnup is approximately 65,000 MWd/tU.

#### Gross Gamma Scan of Limerick Fuel Rod J4



**Figure 3. Gamma scan profile for high-burnup Limerick rod J4.** The vertical lines indicate the locations of the fuel rod segmentation. Red lines are positions of available rod segments.

Just as the HP-RCA measurements are crucial to reducing computational uncertainty, the need for accurate input data related to the operation of the assembly in the reactor is also important. Most information on the design and operating conditions are available from reactor operating records. However, the axial void fraction for the core locations of assembly YJ1433 are not available. Therefore, accurate information from the core simulations performed by the fuel vendor have been requested and are in the process of being compiled for this program.

#### 5. SUMMARY

ORNL has completed the chemical burnup determinations for three HBU fuel specimens and has begun the work for the HP-RCA. All the preparatory work for the HP-RCA has been completed, and four specimens have been dissolved using high-precision hot cell dissolution protocols.

Preliminary depletion simulations have been performed for the eight PWR samples selected for full isotopic analysis. Detailed depletion simulations using assembly-specific design data and sample-specific operating history data have been performed for the three samples subjected to burnup-only measurements.

Work is ongoing; PWR samples are in the lab being dissolved, and BWR samples are being prepared for dissolution.

#### 6. REFERENCES

- 1. K. Banerjee, H. Liljenfeldt, P.L. Miller, J.L. Peterson, R.A. Joseph III, J.B. Clarity, G. Radulescu, R.A. Lefebvre, and J.M. Scaglione, "Consolidating Data on Spent Nuclear Fuel into a Unified Database," *Trans. Am. Nucl. Soc.* 115(1), 279-282, Las Vegas, IL (November 2016).
- 2. Spent Nuclear Fuel Assay Data for Isotopic Validation (State-of-the-art Report), NEA/NSC/WPNCS/DOV(2011)5, Nuclear Energy Agency (2011). <a href="https://www.oecd-nea.org/science/wpncs/ADSNF/SOAR\_final.pdf">https://www.oecd-nea.org/science/wpncs/ADSNF/SOAR\_final.pdf</a>
- 3. I. C. Gauld, G. Ilas and G. Radulescu, *Uncertainties in Predicted Isotopic Compositions for High Burnup PWR Spent Nuclear Fuel*, NUREG/CR-7012, US Nuclear Regulatory Commission Office of Nuclear Regulatory Research (2011). <a href="https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr7012/">www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr7012/</a>
- B. L. Broadhead, M. D. DeHart, J. C. Ryman, J. S. Tang, and C. V. Parks, *Investigation of Nuclide Importance to Functional Requirements Related to Transport and Long-Term Storage of LWR Spent Fuel*, ORNL/TM-12742, Oak Ridge National Laboratory, Oak Ridge, TN (1995). https://info.ornl.gov/sites/publications/Files/Pub57421.pdf
- 5. I. C. Gauld and J. C. Ryman, *Nuclide Importance to Criticality Safety, Decay Heating, and Source Terms Related to Transport and Interim Storage of High-Burnup LWR Fuel*, NUREG/CR-6700, US Nuclear Regulatory Commission Office of Nuclear Regulatory Research (2001). <a href="https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr6700/">https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr6700/</a>
- 6. I. C. Gauld and U. Mertyurek, *Margins for Uncertainty in the Predicted Spent Fuel Isotopic Inventories for BWR Burnup Credit*. NUREG/CR-7251, US Nuclear Regulatory Commission Office of Nuclear Regulatory Research (2018). <a href="https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr7251/">https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr7251/</a>
- 7. G. Radulescu, I. C. Gauld, G. Ilas, and J. C. Wagner, *An Approach for Validating Actinide and Fission Product Burnup Credit Criticality Safety Analyses Isotopic Composition Predictions*, NUREG/CR-7108, U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research (April 2012). www.nrc.gov/docs/ML1211/ML12116A124.pdf
- 8. ASTM E321, Standard Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)
- 9. W. A. Wieselquist, R. A. Lefebvre, and M. A. Jessee, Eds., *SCALE Code System*, ORNL/TM-2005/39, Version 6.2.4, Oak Ridge National Laboratory, Oak Ridge, TN (2020).

## APPENDIX A. PWR SAMPLE INFORMATION

### **List of PWR Isotopic Specimens**

Rod		ng segment range (mm)	Estimated burnup (GWd/MTU)	Sample length	Fuel burnup only	Order in which specimen was processed	Physical location of specimen
30AE14*	2675	2694	60.6	8.9 mm			REDC
30AD05*	2410	2429	58.8	9.2 mm			REDC
3D8E14*	2655	2674	63.5	8.3 mm			REDC
3D8E14*	2303	2322	62.2	12.4 mm			REDC
3F9N05*	2300	2329	57.3	10.3 mm			REDC
3F9N05*	2863	2882	57.8	9.1 mm			REDC
3D8E14*	700	719	61.2		14.9 mm	2 (DONE)	REDC
3A1F05*	2383	2402	53.3	9.8 mm			REDC
30AD05*	1280	1299	59.2	11.7 mm			REDC
3D8E14**	3206	3225	59.1		8.6 mm	1 (DONE)	REDC
6U3K09**	3506	3525	42.5		12.4 mm	3 (DONE)	REDC
Total number of specimens			Q	3			

**Total number of specimens** 

## APPENDIX B. BWR SAMPLE INFORMATION

Design data for the Limerick GE11  $9 \times 9$  assembly YJ1433 are listed in Table B.1. The assembly was irradiated for three operating cycles—5, 6, and 7—and was discharged March 21, 1998. The layout of the GE11 assemblies and the J4 and J6 rod locations is shown in Figure B.1.

Table B.1. Limerick-1 GE11  $9 \times 9$  assembly parameters.

Reactor and assembly data	
Reactor	Limerick Unit 1
Operating pressure (bar)	70
Assembly type	GE11 9 × 9
Fuel assembly pitch (cm)	15.24
Number of fuel rods	74
Number of gadolinium rods	9
Number of water rods	2
Fuel rod pitch (cm)	1.438
Channel bypass gap (cm)	[ ]
Channel thickness (cm)	[ ]
Channel corner radius (cm)	[ ]
Channel material	Zirc4
Channel temperature (K)	560
Active fuel length (cm)	
Fuel rod data	
Pellet radius (cm)	0.471
Pellet material	$\mathrm{UO_2}/\mathrm{Gd_2O_3}\text{-}\mathrm{UO_2}$
Pellet density (g/cm <sup>3</sup> )	10.45
Fuel temperature (K)	1100
Clad inner radius (cm)	0.4878
Clad outer radius (cm)	0.559
Clad material	Zirc2
Clad temperature (K)	560
Water rod data	
Inner radius (cm)	[ ]
Outer radius (cm)	2.489
Water rod material	Zirc2
Water rod temperature (K)	560

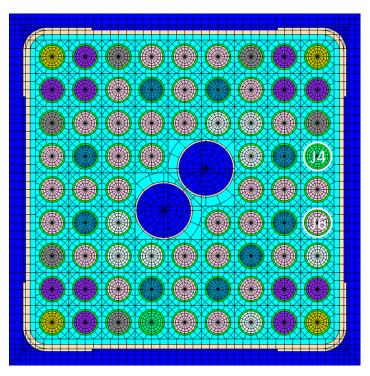


Figure B.1. Limerick GE11 assembly YJ1433 model showing the locations of available fuel rod segments.

## APPENDIX C. ISOTOPES TO BE ANALYZED

Isotope	Separation and detection method	Relative UNCERTAINTY single detector ICPMS (2-sigma )* (best prior practice)	Relative UNCERTAINTY multi-detector ICPMS (2-sigma )* (proposed test method)
<sup>79</sup> Se	PDCA SEP + IDMS with natural selenium	2%	0.5%
<sup>95</sup> Mo	Requires further Study	-	-
<sup>90</sup> Sr	PDCA SEP + IDMS with Sr-86 or natural Sr	5%	0.5%
<sup>99</sup> Tc	Requires further study		-
<sup>101</sup> Ru			0.5%
<sup>106</sup> Ru	PDCA SEP + IDMS with natural Ru	5%	0.5%
<sup>103</sup> Rh			0.5%
<sup>109</sup> Ag	Requires further study	-	-
<sup>125</sup> Sb	DTRA SEP + IDMS with Sb	2%	0.5%
<sup>133</sup> Cs			0.5%
<sup>134</sup> Cs	PDG4 GED + IDMG - 14 - + 1 G		0.5%
<sup>135</sup> Cs	PDCA SEP + IDMS with natural Cs	2%	0.5%
<sup>137</sup> Cs**			0.5%
<sup>143</sup> Nd			0.5%
<sup>145</sup> Nd		2%	0.5%
<sup>146</sup> Nd	Oxalic SEP +IDMS Nd-150		0.5%
<sup>148</sup> Nd**			0.5%
<sup>144</sup> Nd			0.5%
<sup>144</sup> Ce	Oxalic SEP +IDMS Ce-140	2%	1.0%
<sup>147</sup> Pm	Oxalic SEP m/z 147 mass quant using <sup>147</sup> Sm	5%	-
<sup>147</sup> Sm			0.5%
<sup>149</sup> Sm			0.5%
<sup>150</sup> Sm	Oxalic SEP + IDMS <sup>152</sup> Sm	2%	0.5%
<sup>151</sup> Sm			0.5%
<sup>152</sup> Sm			0.5%
<sup>151</sup> Eu			0.5%
<sup>153</sup> Eu	Onalia CED + IDMC 151E	20/	0.5%
<sup>154</sup> Eu	Oxalic SEP + IDMS <sup>151</sup> Eu	2%	0.5%
<sup>155</sup> Eu			0.5%
<sup>155</sup> Gd	Oxalic SEP + IDMS Gd-nat/155Gd	2%	0.5%
<sup>227</sup> Ac	Weighted dilution gamma	5%	-
234U			0.1%
235U**	High annial attention at the All H 222	0.50/20/	0.1%
236U	High precision titration or IDMS with U-233	0.5% or 2%	0.1%
238U**			0.1%

Isotope	Separation and detection method	Relative UNCERTAINTY single detector ICPMS (2-sigma )* (best prior practice)	Relative UNCERTAINTY multi-detector ICPMS (2-sigma )* (proposed test method)
<sup>237</sup> Np	Matrix matched external calibration – weighted dilutions	5%	-
<sup>238</sup> Pu			0.1%
<sup>239</sup> Pu**	IDMS IRMM-86/CRM-130 <sup>239</sup> Pu/ <sup>242</sup> Pu STD		0.1%
<sup>240</sup> Pu**	Alpha ratio for <sup>238</sup> Pu: <sup>239/240</sup> Pu (ASTM protocol)	2%	0.1%
<sup>241</sup> Pu**	– online and offline SEP		0.1%
<sup>242</sup> Pu			0.1%
<sup>241</sup> Am	0. 11. 11. (11.1 GED. IDMG. 11.2014	2%	0.1%
<sup>242</sup> mAm	Oxalic acid actinide SEP + IDMS with <sup>241</sup> Am or <sup>243</sup> Am, <i>verification of 241 using gamma</i>		0.5%
<sup>243</sup> Am	or run, verticulon of 241 using gumma		0.1%
<sup>242</sup> Cm			1.0%
<sup>243</sup> Cm			1.0%
<sup>244</sup> Cm	Oxalic acid actinide SEP + IDMS with in-house	2%	1.0%
<sup>245</sup> Cm	certified Cm-WRM, verification of 242 using 6.11 alpha and 243 using gamma	270	1.0%
<sup>246</sup> Cm			1.0%
<sup>247</sup> Cm			1.0%

<sup>\*</sup>Based on the isotopic concentration of 1  $\mu$ g/g fuel \*\*Isotopes required for the determination of chemical burnup Unable to measure these isotopes with current analytical capabilities:  $^{14}$ C;  $^{36}$ Cl,  $^{221}$ Fr